Concentration dependence of water diffusion in silica glass at water pressure of 50 bar

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Water is the most abundant component in volcanic gas in subduction zones, and its bubble nucleation and growth have strong influences on magmatisms, such as silicate melt properties and a volcanic eruption style. Diffusion of water in silicate melts is one of the controlling factors of the bubble growth in magma, and it is thus important to be clarified to understand the mechanism of volcanisms in subduction zones.

Water diffusivity in silicate melt is known to depend on its own concentration (e.g., Wakabayashi and Tomozawa, 1989; Doremus, 2000; Zhang and Behrens, 2000; Okumura and Nakashima, 2004). However, the diffusion mechanism that is responsible for the observed water concentration dependence has not yet be fully understood. In this study, we carried out diffusion experiments of water in SiO$_2$ glass at 650-850 degree C and water vapor pressure of 50 bar in order to understand the mechanisms of water diffusion in silicate melt. We used SiO$_2$ glass because it allows us to consider only the water concentration dependence of water diffusion without any other compositional effects (e.g., NBO/T).

A SiO$_2$ glass chip was enclosed in a silica glass tube with pure water corresponding to a water vapor pressure of 50 bar at experimental temperatures. The sealed glass tube was heated in a box furnace over the range from 650 to 850 degree C for 25 and 40 hours. Diffusion profiles of $^1$H and $^{30}$Si were measured on the cross section of a run product with the ion microprobe ( Cameca ims-6f at Hokkaido University) using a Cs$^+$ primary beam (20 nA and 20 micron in diameter). Negatively charged ions of $^1$H and $^{30}$Si were counted for 2 sec. and 1 sec. respectively, with a 5-micron step. An electron gun was used for electrostatic charge compensation. Glasses with known water contents were used for conversion of secondary ion counts ($^1$H/$^{30}$Si) to the water content in the samples.

The surface concentration of water in run products ranged from 0.25 to 0.35 wt%. The concentration profiles of water from the surface, especially at the low concentration region, cannot be simply explained by previously proposed diffusion models, where the diffusion coefficient of water is proportional simply to the water concentration or to a relative abundance of molecular water to the total dissolved water and an empirical exponential function of total water content. The concentration profiles obtained in this study show much stronger water-concentration dependence compared with previous studies.

We explain this stronger water concentration dependence of water diffusion in SiO$_2$ glass as follows: (1) The main diffusion species is molecular H$_2$O, and its relative abundance to OH groups decreases with lowering the water content in the glass; (2) The number of pathways available for diffusion is controlled by the number of non-bridging oxygen that is determined by the concentration of OH groups. Because the diffusion coefficient of water is proportional to the above factors (the relative abundance of molecular water and the number of pathways for diffusion) and both depend on the total water content in the glass, the water-concentration dependence of water diffusion is larger than the previous models proposed for rhyolite glasses.

The smaller water-concentration dependence observed for water diffusion in other silicate glasses can be attributed to the little dependence of non-bridging oxygen concentration on water contents because it is controlled extrinsically by other network modifier cations.

Keywords: water, diffusion, silica glass, diffusion model